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Air Pollution Assessment of Cresols

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Air Pollution Assessment of Cresols

J. GORDON

JUNE 1976

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Cresol is a methyl substituted, hydroxylated aromatic hydrocarbon that occurs in three isomeric forms. Cresols are refined from coal tar or petroleum; they can also be synthesized.

Cresols are toxic to both fauna and flora. Poisoning of humans has resulted from inhalation, ingestion, and absorption of cresol through skin and mucous membranes. Cresol affects the central nervous system, the kidneys, liver, lungs, heart, and stomach. Some fatalities and also carcinogenicity have been attributed to cresol.

The principal sources of cresol emissions to the atmosphere are the production of cresols and the production of coke. Adsorption onto activated carbon is the best method for controlling the emission of cresol vapors into the atmosphere.

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I. SUMMARY

Cresol is a methyl-substituted, hydroxylated, aromatic hydrocarbon. Its three isomers are termed ortho-, meta-, and para-cresol. Although o-cresol can be separated from the other two isomers, p-cresol and m-cresol are difficult to separate and they are often marketed as binary mixtures. The term tricresol applies to mixtures of the three isomers, and cresylic acid refers to crude cresol which may also contain other phenolic compounds.

Cresols are toxic to both fauna and flora. Human exposure and poisoning has resulted from inhalation, ingestion, and absorption through skin and mucous membranes. Fatalities have been reported. Cresol affects the central nervous system, the kidneys, liver, lungs, heart, and stomach.

Dose-response studies with various routes of exposure (intravenous, intraperitoneal, skin, subcutaneous, oral) have provided LD_{Lo} and LD_{50} values for various animals (dog, cat, rabbit, guinea pig, rat, and mouse). Carcinogenicity of cresol has been reported by several investigators.

Cresol is generally toxic to microorganisms although it can serve as substrate for certain yeasts and it is a metabolite of certain fungi. The effect of cresol on plants is variable; it is a herbicide, but it also is a dormancy-breaking agent and it has selective chemotherapeutic action against plant neoplasms.

No data on ambient concentrations of cresol in air are available although techniques for sampling air and determining its cresol content have been developed.

The population at risk consists primarily of workers in plants where cresols are purified, synthesized, and processed. Five parts per million has been recommended by the American Conference of Governmental Industrial Hygienists and adopted as a United States Occupational Standard -- i.e., as the maximum time-weighted average (TWA) concentration of cresol to which a worker should be exposed during a 40-hour work week.

Cresol is economically valuable, and consequently it is unlikely that significant quantities of cresol are released into the environment from production and processing plants. Nevertheless, even if a hypothetical 80-million-pound-per-year production facility were to operate with a loss in yield of 10 percent and emission controls that were 98 percent effective, the ambient cresol concentration 500 meters downwind of the plant would be only 0.037 ppm. A person would have to remain at that position for approximately 5400 hours or 225 days in order to inhale an amount of cresol equivalent to that inhaled by a worker who is exposed to the recommended maximum TWA of 5 ppm. A point 500 meters from the plant would be within the perimeter of the facility, and ambient concentrations in adjacent neighborhoods would be lower.

The principal sources of cresol emissions to the atmosphere are the production of cresols and the production of coke. It is estimated

that in the United States as much as 3,393,000 pounds of cresol may be added to the air annually by these processes -- 443,000 pounds during cresol production and 2,950,000 pounds during coke production.

The general population is at risk to cresols from various sources. Saponated cresol is a commonly used household disinfectant. Cresol has been detected in brewed tea; it was carcinogenic. Cresols are present in tobacco smoke (cigarette, cigar, pipe, water pipe, and marijuana), and the carcinogenicity of the cresols isolated from cigarette smoke condensate has been demonstrated. This places at risk not only smokers but also the nonsmoking population which is exposed to cigarette smoke. p-Cresol is a fragrance raw material; its use in flavoring substances is permitted in the United States.

Cresols are derived from coal tar and petroleum by extraction and fractionation. Increasing quantities are now being synthesized from toluene, by methylation of phenol, and by oxidation and cleavage of cymene. Cresols are used in a variety of products including disinfectants, fungicides, bactericides, herbicides, dyes and pigments, antioxidants, surfactants, flotation reagents, resins, plasticizers, and tricresyl phosphate.

A Chemical Safety Data Sheet has been prepared by the Manufacturing Chemists Association; it details the hazards of cresols and specifies protection equipment, preventive measures, and first aid techniques. The odor threshold concentration of cresol is about one-tenth the threshold limit value for adult worker exposure and

cresol has a strong di ·inct odor; therefore, unknown large concentrations of cresol in air are unlikely.

Ambient data on cresol concentrations should be collected in order to provide a sound basis for assessing the problem of cresol as an air pollutant. In addition, inhalation studies are needed in order to determine the toxicity of cresol in air. The significance of cigarette smoke as a source of cresol in air should be evaluated and also the hazard that this presents to nonsmokers as well as smokers. Various cigarette filters should be tested to determine if this source of cresol in air can be eliminated.

The present U. S. standards for permissible levels of cresol in the working environment should be reevaluated in light of the findings from the recommended inhalation studies. This is of particular import because of the discrepancy in the standards adopted in the United States and in the Federal Republic of Germany. The rationale for the lower threshold limit value (TLV) for worker exposure to cresol in Germany should be examined. Similarly, the use of p-cresol in raw materials for flavoring substances is banned in Europe whereas it is permissible in the United States. Consequently, the Food and Drug Administration should reexamine the use of p-cresol and other cresol-containing compounds as food additives. Reevaluation of American standards is considered warranted and advisable.

II. AIR POLLUTION ASSESSMENT REPORT

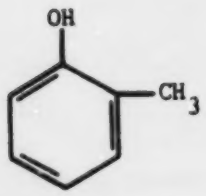
A. PHYSICAL AND CHEMICAL PROPERTIES OF CRESOL

Cresol--also known as hydroxymethylbenzene, methylphenol, and hydroxytoluene--is an aromatic hydrocarbon that is derived from petroleum and coal tar. It has a molecular weight of 108.1 and the formula $CH_3C_6H_4OH$. There are three isomers of cresol (Figure 1)--the ortho, meta, and para forms--which are determined by the relative positions of the hydroxyl and methyl groups on the benzene ring (1).

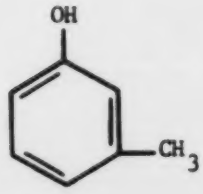
Of the three cresol isomers, o-cresol can be obtained easily in the pure state, but the m- and p-isomers are difficult to separate and they are usually present together in a mixture. Tricresol is the term for a mixture of the three isomers. Crude cresol (commercial cresol) is a mixture of aromatic compounds that contains about 20 percent o-cresol, 40 percent m-cresol, 30 percent p-cresol, and small amounts of phenol and xylenols (2); its boiling point is below 204°C (3). Cresylic acid is the trade designation for commercial mixtures of phenolic materials that boil above the cresol range; by an arbitary standard, 50 percent of the material must boil above 204°C (3).

Cresol has a sweet, tarry odor. At room temperature, it may be liquid or crystalline. It is colorless or yellowish when prepared, but the color darkens on exposure to air or light. Cresol is an oily substance, but it is more dense than water. The physical properties of the ortho-, meta-, and para-cresol isomers and of commercial cresol are presented in Tables I, II, III, and IV. o-Cresol is very

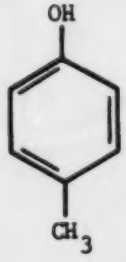
FIGURE 1
THE THREE ISOMERS OF CRESOL



ortho
(o-cresol)



meta
(m-cresol)



para
(p-cresol)

TABLE I
PHYSICAL PROPERTIES OF THE CRESOLS

	Commercial Cresol	o-Cresol	m-Cresol	p-Cresol	Ref.
Melting point, °C	10.9-35.5	30.8	10.9	35.5	1
Boiling point, °C	191-203	190.8	202.8	201.8	1
Density at 20°/4°C	1.030-1.038	1.047	1.034	1.0341	1
n_D^{20}		1.5453	1.5398	1.5395	4
Specific heat (0-20°C), cal/g		0.499	0.479		4
Specific heat (22-45°C), cal/g				0.483	4
Flash point (closed cup), °C	81.1	81.1	94.4	94.4	1
Surface tension at 20°C, dyn/cm		40.3	37.6	39.2	4
Critical density, g/ml		0.374	0.357	0.347	4
Critical pressure, atm		49.4	45.0	50.8	4
Critical temperature, °C		422.3	432.0	426.0	4
Latent heat of fusion, cal/g		30.1		26.3	4
Latent heat of vaporization, cal/g		99.9	104.7	105.1	4
Heat of combustion at constant pressure and 25°C, cal/g		8162	8187	8175	4
Crystallinity		crystals or liquid	liquid	crystals	1
Solubility					
in water, %	1.0	2.5	0.5	1.8	2
in mineral oil, %		2.0	2.5	0.7	2
in alcohol		+	+	+	3
in ether		+	+	+	3
in chloroform		+	+	+	3
Autoignition temperature, °C		598.9	558.9	558.9	1
Lower explosion limit, % at °C	1.35, 148.9°	1.4, 148.9°	1.1, 150.0°	1.1, 150.0°	1

Source: Ref. 1: N. I. Sax, "Dangerous Properties of Industrial Materials," 4th ed., Van Nostrand Reinhold, New York, pp. 579-580, 1975.

Ref. 2: D. W. Fassett and D. D. Irish, Eds., "Toxicology," Volume II in "Industrial Hygiene and Toxicology," (F. A. Patty, Ed.), 2nd ed., Interscience, New York, pp. 1387-1392, 1963.

Ref. 3: A. Rose and E. Rose, "The Condensed Chemical Dictionary," 7th ed., Van Nostrand Reinhold, New York, pp. 257-259, 1966.

Ref. 4: D. McNeil, "Cresols," in "Kirk-Othmer Encyclopedia of Chemical Technology," 2nd ed., Vol. 6, pp. 434-444, Wiley, New York, 1965.

TABLE II
TEMPERATURE-DEPENDENT PROPERTIES OF THE CRESOLS

	Commercial Cresol		o-Cresol		m-Cresol		p-Cresol		Ref.
	Temp, °C	Value	Temp, °C	Value	Temp, °C	Value	Temp, °C	Value	
Saturated vapor pressure, mm Hg	38-53	1	25	0.2453	25	0.1528	25	0.1080	2
			38.2	1	52.0	1	53.0	1	1
			90.6	20	101.3	20	101.8	20	4
			105.2	40	115.9	40	116.4	40	4
			138.58	153.09	149.0	150	145.59	131.39	4
			163.53	349.31			162.72	238.70	4
			172.00	443.55	165.86	299.50			4
			185.19	650.92	190.27	549.23	183.66	457.50	4
					199.00	697.74	197.10	667.77	4
Viscosity, cP			0	39.7	0	84.4	0	98.4	4
			20	9.56	20.2	17.30	20	18.95	4
			40	4.15	40	6.22	40	6.50	4
			120	0.737	123.5	0.820	130	0.801	4
			150	0.504	150	0.580	150	0.615	4
Dielectric constant			25	11.479	25	11.749			4
			30	10.937	30	11.237	35	17.00	4
			58	6.02	58	9.68	58	9.91	4
Solubility, g/100 g H_2O			0	1.3					4
			46.2	2.9	50.8	2.7	30	2.21	4
			104.5	4.5	92.2	4.5	105	5.4	4
			121	6.9	140.4	14.0	138	16.4	4
			155.35	10.93	148.7	29.7	142.6	32.1	4
			169.25	35.51					4
Percent in saturated air			25	0.0323	25	0.0201	25	0.014	2
Density of saturated air (air = 1)			25	1.00089	25	1.00054	25	1.00039	2

Source: Ref. 1: N. I. Sax, "Dangerous Properties of Industrial Materials," 4th ed., Van Nostrand Reinhold, New York, pp. 579-580, 1975.

Ref. 2: D. W. Fassett and D. D. Irish, Eds., "Toxicology," Volume II in "Industrial Hygiene and Toxicology," (F. A. Patty, Ed.), 2nd ed., Interscience, New York, pp. 1387-1392, 1963.

Ref. 4: D. McNeil, "Cresols," in "Kirk-Othmer Encyclopedia of Chemical Technology," 2nd ed., Vol. 6, pp. 434-444, Wiley, New York, 1965.

TABLE III
THERMODYNAMIC FUNCTIONS OF THE CRESOLS*

	o-Cresol	m-Cresol	p-Cresol
At 298.16°K			
heat capacity, C_pm cal/deg	31.15	29.75	29.75
entropy, S°, cal/deg	85.47	85.27	83.09
heat of formation, - H_f, kcal	30.74	31.63	29.97
free energy of formation, G_f, kcal	-8.86	-9.69	-7.67
At 0°K			
heat of formation, H_f, kcal/mole	-25.25	-25.84	-24.21

*Values given for one mole of ideal gas at one-atmosphere pressure

Source: D. McNeil, "Cresols," in "Kirk-Othmer Encyclopedia of Chemical Technology," 2nd ed., Vol. 6, pp. 434-444, Wiley, New York, 1965.

TABLE IV
SPECIFICATIONS FOR COMMERCIAL CRESOLS

	o-Cresol	m-Cresol	p-Cresol	Commercial Cresol	Cresol N.F. XII*	Cresylic Acid**
Composition	varies with grade	varies with grade	varies with grade	by grade; mixtures of the o-, m- and/or p-cresols	isomeric cresols	m-, p-cresols 0-1% 2,4-, 2,5-xylenols 0-3% 2,3-, 3,5-xylenols 10-20% 3,4-xylenol 20-30% C_9 phenols 50-65%
Boiling point, °C	191	203	202	< 204°C for 50%	90% (v): 195-205	220-250
Specific gravity	1.0511	1.034	1.039			
Weight, lbs/gal	8.61	8.66	8.66		8.67	
Technical grades	by melting point (e.g., 25, 29, 30, 30.5°C	95-98%	98% 99% (m.p. ≥ 34°C)	variety, depend e.g., on color, on distillation range, on water, acid or phenol content	N.F. XII	
Solubility alcohol ether chloroform water glycol dilute alkalis	soluble soluble soluble slightly soluble	soluble soluble soluble slightly soluble	soluble soluble soluble slightly soluble		soluble soluble slightly soluble soluble soluble	
Containers	drums tank trucks tank cars	drums	drums tank trucks tank cars			5-, 55-gal drums tank trucks tank cars

*Cresol N.F. XII is one grade of commercial cresol.
**Values given are for a typical commercial cut. By definition, cresylic acid is a mixture of cresols, xylenols, and phenols of which at least 50 percent boil above 204°C.

Source: Data from A. Rose and E. Rose, "The Condensed Chemical Dictionary," 7th ed., Van Nostrand Reinhold, New York, pp. 257-259, 1966.

reactive toward most radicals. It is degraded by HO and by ozone in air, and by organic radicals (RO_2) in water. Its estimated lifetime is less than one day in air and less than 10 days in water. The degradation products are believed to be quinones and hydroxybenzenes (5).

The chemical behavior of the cresols is similar to that of phenol (4). They are mildly acidic, and they form alkali-metal salts which are readily soluble in water. Cresols are readily substituted by electrophilic and nucleophilic reagents. Under acidic conditions, cresols are readily alkylated with alkyl halides or alkenes. For example, reaction with isobutylene yields butylcresol whereas reaction with dimethyl sulfate yields ethers (methoxytoluenes or methylanisoles) (4). Although they are acidic, cresols can form esters with weak acids and with acid chlorides. Thus, reaction with $POCl_3$ forms tricresyl phosphate, $(CH_3C_6H_4)_3PO_4$.

The product of cresol oxidation depends on the oxidizing agent used. It may be, for example, hydroxybenzoic acid, quinones, or hydroquinones. The product of catalytic reduction is a function of the catalyst used (e.g., nickel, noble metals) and the conditions of the reaction; it could be methylcyclohexanol and/or methylcyclohexanone. When cresols in the vapor phase are treated with hydrogen over mixed oxide catalysts, they are converted more or less completely to toluene; under sufficiently drastic conditions, the product is benzene (4).

Cresols also condense with formaldehyde (4). At room temperature in dilute alkali, the condensation product is methylhydroxylbenzyl alcohol. Under acidic or neutral conditions and higher temperatures, thermosetting resins are formed.

When cresol vapor is exposed to heat or flame, there is a slight danger of explosion. The lower explosion limits for commercial cresol and o-cresol are 1.35 percent and 1.4 percent, respectively, at 148.9°C. These limits are similar to values for m- and p-cresol, each of which has a lower explosion limit of 1.1 percent at 150°C (1).

Cresols are slightly flammable. The closed cup flash point of commercial cresol and o-cresol is 81.1°C and that of m- and p-cresol is 94.4°C (1). Carbon dioxide, dry chemicals, carbon tetrachloride, and foam are recommended to fight cresol fires.

As disaster hazards, cresols are classified as dangerous because, when heated to decomposition, they emit highly toxic fumes. Furthermore, they can react vigorously with oxidizing materials (1).

B. BIOLOGICAL EFFECTS OF CRESOL

1. Humans

Cresol is a cytoplasmic poison, and therefore it affects the entire organism, but especially the central nervous system, liver, and kidneys (6). The lungs, pancreas, and spleen may also be injured (2). Acute exposures of all types may cause muscular weakness, gastroenteric disturbances, severe depression, collapse, and death. Repeated exposures may result in digestive disturbances, damage to the liver and kidneys, and skin eruptions (2). The toxic effects of cresols on humans are tabulated in Table V.

Central nervous system disorders induced by cresol include dizziness, headaches, delirium, hallucinations, and confusion in adults and convulsions in children (6). Pathologic findings include cerebral edema (12). Liver involvement was indicated by turbidity, inflammatory reactions, and fatty degeneration (2). Parenchymatous and hemorrhagic nephritis* were found in the kidney as well as lower nephron nephrosis (2, 10). Hyperemia,** emphysema, edema, and bronchopneumonia were reported (2). The heart was affected also; the myocardium*** was degenerated and there were small hemorrhages in the epicardium**** and endocardium***** (2). There was necrosis of mucous membranes (12).

*Inflammation of the kidney.
**Congestion with an usual amount of blood.
***Muscular tissue of the heart.
****Inner part of the outermost layer of the heart wall.
*****Membrane lining of the inner surface and cavities of the heart.

TABLE V
TOXIC EFFECTS OF CRESOLS ON HUMANS

Age	Mode of Exposure	Dose	Type of Cresol	Effects	Result	Ref.
30 yrs	inhalation	8 hrs	cresol	considerable pulmonary congestion heavy phlegm abnormal lung X-ray	recovery	6
60 yrs	ingestion	250 ml	cresol	congested liver very congested stomach hemorrhaged pancreas	death in 36 hrs.	7
48 yrs	ingestion	250 ml	conc cresol	oral-pharyngeal burns pulmonary congestion delayed severe hemolytic anemia	slow recovery (6 wks.)	6
62 yrs	ingestion	150 ml	lysol*	collapse confusion elevated serum total phenols	slow recovery	8
63 yrs	ingestion	unk amt	lysol	vomiting unconsciousness distressed breathing superficial burns on shoulder blistering around mouth edema of the pharynx and glottis cyanosis collapse of right lung	recovery (2 wks.)	9
35 yrs	ingestion	20 ml	cresol	collapse acetonuria black urine	recovery	6
	ingestion	a cupful	cresol		recovery	6
	ingestion	a little	cresol		recovery	6
	ingestion	a little	cresol		recovery	6
	ingestion	a little	cresol		recovery	6
2 yrs	ingestion	a sip	cresol	vomiting	recovery	6
3 yrs	ingestion	a sip	cresol	bloody urine 2 days later jaundice	recovery	6
5 yrs	ingestion	a sip	cresol	vomiting	recovery	6
7 yrs	ingestion	a sip	cresol	vomiting	recovery	6
26 yrs	vaginal douche		lysol	renal shutdown lower nephron nephrosis pulmonary edema tubular necrosis	death on 6th day	10
32 yrs	vaginal douche		lysol	renal shutdown lower nephron nephrosis	recovery	10
3 yrs	skin		cresol	hypotension, hyperpnea, convulsions, coma, pulmonary edema	death	11
1.5 yrs	skin		cresol	extensive burns of head, lower abdomen, perineum	slow healing of burns	11

*Lysol: 50% in saponated solution.

Cresols have been rated as moderately toxic to humans; see Table VI (1). The specific effects and symptoms of cresol poisoning depend on the type of exposure. Cresol may be absorbed through the skin and through the mucous membranes of the gastroenteric and respiratory tracts. In general, cresols are caustic and corrosive and consequently cause burns of the tissues which they contact--be it skin or mucosa (1, 2, 6). Application to the skin causes burns and/or blistering (6, 9), and repeated exposures may cause dermatitis (1, 2, 13). Ingestion of cresol often results in burns and edema of the mouth, glottis, pharnyx, esophagus, and stomach (2, 6, 9).

Taste and smell would tend to limit the ingestion of appreciable quantities of cresol. Nevertheless, ingestion of cresol has caused numerous cases of cresol poisoning (6, 7, 8, 9) and one reported fatality (7). Absorption of cresol through mucous membranes (10) and through the skin (11) has also had fatal consequences.

Jouglard et al. (6) reported that, at the Marseille Poison Center during the years 1968 to 1970, the frequency of cresol poisoning was slightly more than 1:1000 with a frequency of 1:2000 for serious cases. In children, poisoning resulted from accidental ingestion of cresol whereas in most adults ingestion was deliberate. Symptoms of cresol poisoning by ingestion included vomiting, abdominal distress, diarrhea, a characteristic breath odor, acetonuria,* black or bloody urine, jaundice, and anemia. One adult was poisoned by accidental

*Acetone and diacetic bodies in the urine.

TABLE VI
TOXIC HAZARD RATINGS OF CRESOLS*

	Allergen	Ingestion	Inhalation	Irritant	Skin Absorption
Acute local	Slight	Moderate	Moderate	Moderate	
Acute systemic		Moderate	Moderate		Moderate
Chronic local	Slight			High	
Chronic systemic		Moderate	Moderate		Moderate

*Slight: Causes readily reversible changes which disappear after end of exposure.
Moderate: May involve both irreversible and reversible changes that are not severe enough to cause death or permanent injury.
High: May cause death or permanent injury after any short exposure to small quantities.

Source: Sax, N. I., "Dangerous Properties of Industrial Materials," 4th ed., Van Nostrand Reinhold, New York, pp. 579-580, 1975.

inhalation of cresol fumes when cresol dripped onto his pillow all night; he suffered serious pulmonary congestion with heavy phlegm.

Cresol poisoning in two women who used concentrated lysol* vaginal douches to induce abortion was caused by absorption of cresol through mucous membranes (10). Both women suffered renal shutdown within 24 hours, and lower nephron nephrosis** was diagnosed. One patient recovered. The other patient died on the sixth day; the postmortem revealed pulmonary edema and tubular necrosis.***

An 18-month-old child suffered extensive burns of the head, face, lower abdomen, and perineum when Cresylene Vaporising Liquid (Cresol BP) was poured over her (11). The burns healed slowly. A similar incident was fatal to a 3-year-old child. Arthur (11) reported that the potential fatal consquences of absorption of cresol through the skin are hypotension, hyperpnea,**** convulsions, coma, and pulmonary edema.

Fairhall reported three cases of toxic effects from industrial exposure to cresol. In one, an irritant sawdust was produced by the sawing of wood painted with crude creosote (which contains cresol) (14). In another, workers in a plant, where an artificial resin that contained cresol was being manufactured, complained of headache,

*Fifty percent cresol with linseed oil soap.
**Degenerative changes in the kidneys without the occurrence of inflammation.
***Death of tissue.
****Increased respiratory rate.

tremor, and vomiting (15). In the third, a cresol-containing spray caused keratitis;* when droplets entered the eye they caused intense irritation and great pain, and small gray spots developed on the cornea which impaired vision (16).

It has been reported that coal tar and coal tar products are carcinogenic (17). Workers exposed for a long period to coal tar have shown a higher than normal incidence of cancer of the skin.

Wodyka reported the case of a man who became hoarse when he started working in a petroleum refinery (18). After working several decades with cresol, dichlorooctane, and chromic acid, he developed squamous cell carcinoma of the true vocal cords.

Cresols usually do not constitute an acute inhalation hazard (2, 6). Their vapor pressure is low at ambient temperatures, and they have a characteristic sweet and tarry odor that is noticeable below hazardous levels. No data are available on the toxicity of cresol vapors to humans (2).

2. Animals

The toxicity of cresol varies with the isomer, the organism, and the mode of exposure (Table VII). The relative toxicity of the three isomers is ortho > para > meta in the cat and rat and ortho = para > meta in the mouse. The relative toxicity in the rabbit, however, is para > ortho > meta when cresol was applied to the skin or administered orally and ortho = para > meta when cresol

*Inflammation of the cornea.

TABLE VII
TOXIC EFFECTS OF CRESOLS ON ANIMALS

Organism	Mode of Exposure	Dose	Type of Cresol	Effects	Results*	Ref.
Dog	intravenous	80 mg/kg	o-cresol		LD_{Lo}	19
Cat	subcutaneous	55 mg/kg	o-cresol		LD_{Lo}	19
Cat	subcutaneous	180 mg/kg	m-cresol		LD_{Lo}	19
Cat	subcutaneous	80 mg/kg	p-cresol		LD_{Lo}	19
Rabbit	skin	1100 mg/kg	o-cresol		LD_{50}	19
Rabbit	skin	2050 mg/kg	m-cresol		LD_{50}	19
Rabbit	skin	301 mg/kg	p-cresol		LD_{50}	19
Rabbit	oral	940 mg/kg	o-cresol		LD_{Lo}	19
Rabbit	oral	1400 mg/kg	m-cresol		LD_{Lo}	19
Rabbit	oral	620 mg/kg	p-cresol		LD_{Lo}	19
Rabbit	intravenous	180 mg/kg	o-cresol		LD_{Lo}	19
Rabbit	intravenous	280 mg/kg	m-cresol		LD_{Lo}	19
Rabbit	intravenous	180 mg/kg	p-cresol		LD_{Lo}	19
Rat	skin	620 mg/kg	o-cresol		LD_{50}	20
Rat	skin	1100 mg/kg	m-cresol		LD_{50}	20
Rat	skin	750 mg/kg	p-cresol		LD_{50}	20
Rat	oral	1454 mg/kg	cresol		LD_{50}	19
Rat	oral	121 mg/kg	o-cresol		LD_{50}	19
Rat	oral	242 mg/kg	m-cresol		LD_{50}	19
Rat	oral	207 mg/kg	p-cresol		LD_{50}	19
Guinea pig	intraperitoneal	100 mg/kg	p-cresol		LD_{Lo}	19
Mouse	inhalation		cresol	hematuria, convulsions		
Mouse	oral	861 mg/kg	cresol		LD_{50}	19
Mouse	oral	344 mg/kg	o-cresol		LD_{50}	20
Mouse	oral	828 mg/kg	m-cresol		LD_{50}	20
Mouse	oral	344 mg/kg	p-cresol		LD_{50}	20
Mouse	unknown	140 mg/kg	p-cresol		LD_{50}	19
Mouse	skin	1000 mg/kg	cresol		LD_{50}	19
Mouse (agouti CBA/J)	skin	0.5% in acetone, 3X/wk for 3 wks	p-cresol	permanent hair depigmentation		27
Mouse (black)	skin	0.5% in acetone, 3X/wk for 3 wks	p-cresol	permanent hair depigmentation		27

*LD_{Lo}: lowest published lethal dose
LD_{50}: lethal dose giving 50 percent kill.

was administered intravenously. The LD_{50}* data for the least toxic and the most toxic isomers differed by a factor that was as small as 1.8 (cutaneous, rat) and as large as 6.8 (cutaneous, rabbit). With the LD_{Lo}** data, these factors were 1.6 (intravenous, rabbit) and 3.3 (subcutaneous, cat), respectively.

Rabbits are more sensitive than rats or mice to cresols applied cutaneously, but rabbits are least sensitive to cresols administered orally (19, 20). In both these experiments, rats were more sensitive than mice (19, 20). Cresols are most toxic to rabbits when they are administered intravenously; skin applications are more toxic than oral doses (19). In both rats and mice, oral doses are more toxic than skin applications (19, 20).

The only reported cresol inhalation studies were those conducted by Uzhdavini and co-workers (20, 21). Unfortunately, English translations of the articles (from the Russian) are not available. The English abstract of one report (20) cites the effects of inhalation studies on mice, i.e., hematuria*** and convulsions; no English abstract of the other article (21) was available.

Kaiser reported that o-, m-, and p-cresols can be distinguised in different types of black fermented tea (22). He tested brewed tea for carcinogenicity by applying it to the necks of Swiss mice on alternate days for 55 paintings; the initiator, 1 percent 3,4-benzopyrene

*Lethal dose giving 50 percent kill.
**Lowest published lethal dose.
***Blood in the urine.

in acetone, was painted on the neck one time. Of the experimental mice, 40 percent developed epithelial cell carcinoma and 60 percent had precarcinogenic or carcinogenic stages of squamous cell tumors. Among the control animals, which had received initiator only, there were no pathologic lesions.

Boutwell and Bosch reported that the ortho, meta, and para isomers of cresol in tobacco smoke are all active tumor-promoting agents (23). Similarly, Zamfir noted that all three isomers have significant cocarcinogenicity (24). On the other hand, Bock et al. had inconsistent results when they tested the steam distillate of the weak acid fraction of cigarette smoke condensate (CSC) (i.e., the fraction containing volatile phenols) for carcinogenicity; they concluded therefore that this fraction "probably does not contribute importantly to the carcinogenicity of CSC for mouse skin" (25).

Other in vivo animal experiments with cresol include those by Shelley (26, 27), who determined that p-cresol was the active ingredient in laundry ink that caused permanent depigmentation of hair in mice. p-Cresol was also identified as the principal phenolic compound in pea haulm silage effluent (28); at a concentration of 20 mg/100 ml, p-cresol caused onset of an excited state in goldfish (Carassius auratus) in 0.25 minutes and death in 145 minutes. p-Cresol administered orally to rats at a dose of 0.0001 mg/kg per day for eight months induced abnormalities in nervous system functions (29).

In vitro experiments on the effects of cresols include those by Lunde and co-workers on isolated, perfused rabbit lung (30, 31). These investigators found that cresols inhibit vasoconstriction,* that which developed naturally as well as that which was induced by arterially injected ATP. *m*-Cresol had only a slight effect, but the inhibitory effect was much greater with the ortho and para isomers of cresol. This finding on the relative toxicity of the three isomers corresponds with the LD_{Lo} and LD_{50} data.

Johnson studied the effects of *m*-cresol on toad embryos at the neural tube stage of development (32). At 1000 ppm, *m*-cresol caused death within three minutes; at concentrations of 10 to 100 ppm, the survival rate was greater than 50 percent. After 48 hours, embryo length had decreased with *m*-cresol at 10 to 60 ppm. Concentrations of 20 to 80 ppm caused two developmental abnormalities: edema and a flexion** in the tail.

The toxic effects of cresol on nematodes is utilized in a commercial nematocide preparation (33) that contains 30 percent cresol. Cresol is also effective as an insecticide (34).

3. Microorganisms

Cresols are generally toxic to microorganisms. However, they can serve as substrates for certain organisms (*e.g.*, yeasts) (35, 36), and they are metabolites of other organisms (*e.g.*, the fungus *Valsa friesii*) (37).

*Constriction of blood vessels.
**Bending.

Kihara et al. reported that cresol has an inhibitory effect on mycoplasma which increases with increasing concentration (38). Sellers found that cresol slowly inactivates foot-and-mouth disease virus (39). p-Cresol at a concentration of 0.6 percent completely inactivates fixed rabies virus in vitro (40). Cresols are used as fungicide (41, 42) and as bactericide (43, 44, 45).

For a discussion of the effects of cresols on crown gall tumors incited by Agrobacterium tumefaciens and on olive knot tumors incited by Pseudomonas savastanoi, see Section B.4.

4. Plants

Shimizu and Ueki reported that o-cresol is an effective agent for breaking dormancy in barnyard grass seeds, whereas m-cresol and p-cresol are without effect (46). p-Cresol was weakly stimulatory on rhizogenesis in Jerusalem artichoke root tissue cultures in the presence of indole-3-acetic acid (47).

Janssen studied the effect of cresols on the pH optimum of indoleacetic acid oxidase that was derived from various plant organs. With pea root oxidase and m-cresol or o-cresol as cofactor, an increase in cresol concentration shifted with pH optimum to a lower pH value (48). With indoleacetic acid oxidase preparations from pea epicotyl, from cucumber root, hypocotyl and cotyledon, and from oat root and coleoptile, the oxidase pH optimum was 1 to 2 pH units lower with p-cresol as the cofactor than with m-cresol (49).

Schroth and Hildebrand reported that the cresol isomers were moderately selective in destroying plant neoplasms without adversely

affecting contiguous healthy tissues (50). When cresols were applied in a 1.5 percent solution (v/v) in paraffin oil, they destroyed 60 to 80 percent of the tumors. The investigators developed an oil-water emulsion which contained m-cresol as one of the active ingredients. The efficacy of this preparation was demonstrated in field tests against crown gall tumors (incited by Agrobacterium tumefaciens) of almond, peach, apricot, plum, cherry, and pear as well as against olive knot tumors (incited by Pseudomonas savastanoi).

C. AMBIENT DATA, POPULATION AT RISK, AND MEASUREMENT TECHNOLOGY

No data on ambient concentrations of cresols in air were identified, and it could not be ascertained whether any such data have ever been reported.

Cresols have a sweet, tarry odor which has a low atmospheric odor detection threshold (o-cresol: 0.65 ppm; m-cresol: 0.68 ppm) and also a low recognition threshold in air (p-cresol: 1.00 ppb) (51). Consequently, since the threshold limit value (TLV) for cresols is 5 ppm for adult worker exposure (8 hours/day, 5 days/week) as established by the American Conference of Governmental Industrial Hygienists (52, 53), the presence of cresol vapor is evidenced by its odor at concentrations approximately one-tenth that of the TLV. Thus, odor should provide ample warning of the presence of cresol in the air.

The segment of the population primarily at risk of exposure to cresols in air consists of those workers involved in the production of cresols by purification or by synthesis as well as those involved in the manufacture of other chemicals for which cresol is a starting material or an intermediate. Cresols are economically valuable so there is little likelihood that major emissions are produced from industrial facilities during production and manufacture.

In order to estimate the risk generated by a production facility, we can assume a theoretical plant that releases cresol through ventilation and exhaust of internal air. Atmospheric dispersion would distribute the gaseous emissions from this plant downwind.

Hypothetical plant conditions can be inserted in a Gaussian plume equation* (54). The basic diffusion equation should be modified, however, because of the effect of the plant itself on the flow of air. Mechanical turbulence in the wake of a building tends to produce aerodynamic downwash that results in fairly rapid diffusion of a gaseous emission down to the ground. This region of disturbed flow extends downwind a distance equal to several times the height of the building. Although this situation does not result in a Gaussian distributed plume, it is proposed that a modification of the usual formula can still be used. The turbulent mixing in the wake of the building is assumed to be distributed uniformly in the vertical

*Ground level downwind concentrations resulting from a point source are predicted by the following equation:

$$\chi(x,0,0;H) = \frac{Q}{\sigma_y \sigma_z u} \exp -1/2 \left(\frac{H}{\sigma_z}\right)^2$$

where:
- Q = uniform emission rate (grams/sec)
- u = mean wind speed affecting the plume (meters/sec)
- H = effective stack height, that is sum of stack height plus plume rise (meters)
- σ_y = horizontal dispersion coefficient evaluated in terms of downwind distance to the point for which the concentration is being computed (meters)
- σ_z = vertical dispersion coefficient evaluated in terms of downwind distance to the point for which the concentration is being computed (meters)
- $\chi(x,0,0;H)$ = the concentration in grams/meter at a receptor located at ground level, x meters directly downwind from an emission source having an effective stack height of H meters.

direction, analogous to the situation where a plume is trapped below an inversion layer. A limited vertical mixing height can be modeled using the equation:

$$X(x,0,0;H) = \frac{Q}{\pi \sigma_y u\ (.8L)}$$

where L is the limit of the mixing depth. In the case of an isolated rectangular building, it is assumed that L equals 1.5 times the building height.

In addition, it is assumed that there is a horizontal wind turbulence which results in an initial horizontal plume spread equal to the width of the building normal to the wind direction. This is analogous to an area source emission where the area equals the building top. This can be modeled using a further modification of the basic Gaussian diffusion equation. A virtual point source is assumed upwind from the building at a distance where the plume spread (for the given stability conditions) would equal the crosswind width of the building. The modified diffusion equation is therefore:

$$X(x,0,0;H) = \frac{Q}{\pi \sigma'_y u\ (1.2h)}$$

where h is the building height and σ'_y is based on the downwind distance to the receptor point plus the upwind distance to the virtual point source.

Generally speaking, cresol is produced from petroleum tar acids in pressure-tight reaction and separation vessels. Thus atmospheric release should be virtually nil. However, as a conservative approximation, it is assumed that a hypothetical plant producing 80 million pounds of cresol annually experiences a 10 percent loss of yield of which 98 percent is recovered by emission control techniques. The production facility is assumed to be 50 feet (15 meters) high and 100 feet (30 meters) wide. In this hypothetical example, the ambient cresol concentration 500 meters downwind of the plant is estimated to be 0.163 mg/m^3 or 0.037 ppm.* A downwind distance of 500 meters was chosen to represent a point within the perimeter of the production facility. Ambient concentrations in surrounding neighborhoods could be expected to be significantly lower.

Under these assumed conditions, a person would have to remain positioned 500 meters downwind of the facility for approximately 5400 hours or 225 days in order to inhale an amount equivalent to that inhaled by a worker who is exposed to the maximum time-weighted average (5 ppm) during a 40-hour work week that is recommended by the American Conference of Governmental Industrial Hygienists (52, 53). Consequently, the population living in the vicinity of these plants is at low risk of exposure to cresols.

*Additional assumptions; (1) cresol is nonreactive; (2) atmospheric stability class is neutral; and (3) wind speed is 6 meters/second.

The presence of cresols in various substances places at risk those workers who process and/or are exposed to these substances as well as the general population. Three such substances are tea, fragrances, and cigarette and marijuana smoke.

Kaiser reported that he detected the three cresol isomers in fermented tea (22). A large segment of the population is exposed to cresols in tea because tea is man's most widely consumed beverage other than water (22); furthermore, the brewing of tea would facilitate volatilization of the cresols.

p-Cresol is found in a score of essential oils including ylang ylang and oil of jasmine, and it has been used in fragrances since the 1930s (55). It is incorporated into foods, soap, detergent, creams, lotions, and perfume; the concentrations of p-cresol in these products are listed in Table VIII (55). Opdyke also reported that p-cresol is approved by the U.S. Food and Drug Administration for food use (55). However, the Council of Europe declared p-cresol an "artificial flavouring substance not admissable at present" (56). The general American population is thus exposed to the p-cresol present in a wide variety of products.

m-Cresol has been reported in tobacco leaves (57, 58, 59), and all three isomers of cresol were detected in cigarette smoke (23, 24, 57-61), in cigar smoke (57, 58, 59), in pipe tobacco smoke (57, 58), and in water-pipe tobacco smoke (57). Workers in tobacco research who are exposed to the cigarette smoke generated by the smoking machines as well as

TABLE VIII

CONCENTRATION OF p-CRESOL, USED FOR FRAGRANCE, IN VARIOUS PRODUCTS

PRODUCT	CONCENTRATION, %	
	AVERAGE	MAXIMUM
Soap	0.005	0.2
Detergent	0.001	0.02
Creams, lotions	0.001	0.005
Perfume	0.04	0.4

Source: D. L. J. Opdyke, "Fragrance raw materials monographs: p-cresol," Food and Cosmetics Toxicology, 12, 389-390 (1974).

those who handle and analyze the cigarette smoke condensate experience a potential health risk. Persons who smoke are also at risk, primarily as the result of inhalation during smoking. Nonsmokers in the general population are also exposed to the cresols in tobacco smoke although their risk is probably not great because cresol vapor is 3.72 times heavier than air (62) and consequently it settles rapidly.

The level of cresols in tobacco smoke varies with the strain of tobacco (61), the type of tobacco blend (e.g., Virginia, American, Turkish) (59), and the type of filter (e.g., cellulose acetate filters selectively remove cresols from tobacco smoke) (60). In addition, various chemical additives to the tobacco affect the cresol content of the tobacco smoke (61). The average amounts of cresol emitted per cigarette have been reported as 21, 16, and 37 micrograms for the ortho, meta, and para isomers respectively (60) and also as 22, 50, and 50 micrograms respectively (24); the ratios of the three isomers in tobacco smoke were reported as 1.0/0.9/1.1 (59). Since different factors affect the cresol content of tobacco smoke, such figures are quite variable; nevertheless, they do give some indication of the quantities involved.

The ortho and para isomers of cresol were identified in marijuana smoke (63); their combined content was 339 micrograms per marijuana cigarette (which is approximately five-fold the o- and p-cresol content of a regular cigarette). The presence of m-cresol in marijuana smoke is probable but not confirmed.

The greatest risk to the general population is that presented by the commercial cresol solutions which are used in many households as disinfectants. There is danger of accidental or intentional ingestion of cresol (6, 7, 8, 9) and of accidental poisoning by inhalation (6). Open and/or leaking bottles of cresol could contribute significant quantities of cresol vapor to the air in closed rooms over a period of time.

The chemical properties of cresols provide the means for distinguishing them from chemically similar phenol and for identifying each of the isomers. Thus, for example, Fairhall (14) reported that, whereas phenol is soluble in five parts of sodium hydroxide, cresols are not. Reaction of cresols with ferric ion gives a blue-violet color which is fairly permanent with m-cresol. With the ortho and para isomers, however, the solutions become turbid. They can be identified by their reactions with strong picric acid solutions in alcohol: o-cresol forms orange or yellow needles of picrate whereas p-cresol picrate is unstable and no precipitate is formed.

An early method for determining cresols in air was that developed by Maevskaya (64, 65). The technique involved collecting a large air sample in a flask that contained sodium hydroxide solution. Diazotized p-nitroaniline reagent was used, and the cresol was determined colorimetrically by comparison with cresol standards.

Another method involved absorption of atmospheric cresol in spectrograde alcohol and direct determination by ultraviolet spectrophotometry (2).

Methodology and instrumentation have changed radically in the intervening years, and today accepted procedures for qualitative and quantitative identification of the cresols include thin-layer chromatography (59, 66), gas chromatography (63, 67) with flame ionization detector and mass spectroscopy (63). Hrivnak and Macak reported average deviations of 1.2 percent when peak heights in gas chromatography were used to determine quantities (67).

The Michigan Department of Public Health has recommended a procedure for sampling and analysis of air for cresol content (68, 69). A fritted bubbler should be used to collect the air samples at a sampling rate of 0.05 cubic feet per minute. The minimum sampling volume is one cubic foot. Aqueous alkali constitutes a suitable collecting medium. The suggested analytical method is colorimetry.

D. SOURCES OF CRESOL

The principal sources of cresol emissions are facilities for the production of cresol, facilities in which cresol is used in the manufacture of other substances, coke ovens, and the disposal of cresol-containing products and their containers. Estimated emissions of cresols from cresol production facilities and from coke ovens are presented in Table IX. Emissions from the other sources could not be estimated because of lack of data.

1. Production of Cresols

For many years, the only sources of cresol were coal tar (from coke ovens) and gas works tar (74). Then, after World War II, petroleum refineries began to recover cresylics from the spent caustic liquor that had been used to wash petroleum distillates, and petroleum-based cresols were marketed. Today, cresols are also being synthesized--by sulfonation and hydrolysis of toluene, by methylation of phenol, and by oxidation and cleavage of cymene--and synthetic cresols constitute an increasingly larger proportion of total production (74, 75).

Production of cresols by refining from coal tar and petroleum amounted to 115.4 million pounds in the United States in 1973 (71). Production and sales data are listed in Table X. The major United States producers of cresols and the raw materials used at the various plants are listed in Table XI (71, 76), and the locations of these facilities are indicated in Figure 2. Because of the complexity of cresylics and the three isomers of cresol, detailed information is not available about the volume of cresol production at each facility.

TABLE IX

ESTIMATED ANNUAL CRESOL EMISSIONS IN THE UNITED STATES

SOURCE	U.S. PRODUCTION* (yr)	(lbs)	EMISSIONS** (lbs)	RECEIVING MEDIUM
Production of cresol from coal tar and petroleum	1973	115,436,000	230,900 127,000	air water
Synthesis of p-cresol	1972	106,000,000	212,000 116,600	air water
Coke ovens	1974	62,256,066 tons	2,950,000	air

*Production data for 1972 from Ref. 70, for 1973 from Ref. 71, for 1974 from Ref. 72.

**Estimations of emissions to the air are based on an assumed 10 percent loss in yield and pollution control devices that are 98 percent effective.
Estimations of emissions to the water are based on the observed 0.11 percent loss into waste water during synthesis of p-cresol from toluene (70).
Estimations of emissions from coke ovens are based on the estimated hydrocarbon emissions from coke ovens with no emission controls (73).

TABLE X

PRODUCTION AND SALES OF COAL TAR- AND PETROLEUM-DERIVED CRESOLS AND CRESYLICS IN THE UNITED STATES IN 1973

SUBSTANCE	PRODUCTION (lbs)	SALES (lbs)
o-Cresol	24,741,000	24,053,000
(m, p)-Cresol	31,377,000	
All other cresols*	59,318,000	102,972,000
Cresols, total**	115,436,000	127,025,000
Cresylic acid, refined**	57,524,000	64,819,000

*Includes (o, m, p)-cresol from coal tar as well as some m-cresol and p-cresol.

**Includes data for coke ovens and gas-retort ovens as well as data for tar and petroleum refineries and other producers.

Source: U.S. International Trade Commission, "Synthetic Organic Chemicals, U.S. Production and Sales, 1973," ITC Publication 728, Washington, D.C., p. 22, 1975.

TABLE XI
UNITED STATES PRODUCERS OF COAL TAR- AND PETROLEUM-DERIVED CRESOLS

Product	Company	Division	Location	Raw Material
m-Cresol	Koppers	Organic Materials	Oil City, Pa.	
o-Cresol	Continental Oil	Conoco Chemicals, Pitt-Consol Chemicals	Newark, N.J.	
	Koppers	Organic Materials	Follansbee, W. Va.	coal tar
	Merichem		Houston, Tex.	petroleum
	Productol Chemical		Santa Fe Springs, Calif.	petroleum
	Stimson Lumber	Northwest Petrochemical	Anacortes, Wash.	petroleum
p-Cresol	Americal Cyanamid	Organic Chemicals	Bound Brook, N.J.	
	Sherwin-Williams	Sherwin-Williams Chemicals	Chicago, Ill.	
(m,p), Cresol	Koppers	Organic Materials	Follansbee, W. Va.	coal tar
	Merichem		Houston, Tex.	petroleum
	Productol Chemical		Santa Fe Springs, Calif.	petroleum
	Stimson Lumber	Northwest Petrochemical	Anacortes, Wash.	petroleum
(o,m,p)-Cresol	Koppers	Organic Materials	Follansbee, W. Va.	coal tar
	Merichem		Houston, Tex.	
	Productol Chemical		Santa Fe Springs, Calif.	
Cresols, unspecified	Crowley Tar Products		Houston, Tex.	
Cresylic acid	Crowley Tar Products		Houston, Tex.	coal tar
	Koppers	Organic Materials	Follansbee, W. Va.	coal tar
	Merichem		Houston, Tex.	petroleum
	Mobil Oil	North American	Beaumont, Tex.	petroleum
			Paulsboro, N. J.	petroleum
	Productol Chemical		Santa Fe Springs, Calif.	petroleum
	Stimson Lumber	Northwest Petrochemical	Anacortes, Wash.	petroleum
	United States Steel	USS Chemicals	Clairton, Pa.	coal tar

Source: Data from "1975 Directory of Chemical Producers, United States of America," Chemical Information Services, Stanford Research Institute, Menlo Park, California, 1975; and from "Synthetic Organic Chemicals, U.S. Production and Sales, 1973," ITC Publication 728, U.S. International Trade Commission, Washington, D.C., 1975.

FIGURE 2
LOCATION OF PLANTS THAT PRODUCE CRESOL FROM COAL TAR AND PETROLEUM

Cresols are synthesized by three different methods. These are sulfonation and hydrolysis of toluene, methylation of phenol, and oxidation and cleavage of cymene. In 1972, United States capacity for synthesis of p-cresol was 106 million pounds--10 million pounds from toluene, 90 million pounds from phenol, and 6 million pounds from cymene (70). The producing facilities, their locations, capacities, and the synthesis process used at each are listed in Table XII, and the locations of these plants are indicated in Figure 3.

a. Production of Cresol from Coal Tar

Coal tar constitutes a main source of cresols in the United States and the sole natural source in Britain and Europe (4). Coke-oven tars contain 1 to 7 percent tar acids (average, about 3 percent) with 1.04 percent cresols. Gas-works tar, derived from the carbonization of bituminous coal in continuous vertical retorts, have an average tar acid content of 15 percent with 3.44 percent cresols. The tar acid content of low-temperature tars may be as high as 25 percent with 3.33 percent cresols. See Table XIII (4) for the cresol content of different coal tars.

The coal tar is distilled, and a fraction known as middle oil, creosote oil, or tar-acid oil is collected. From this fraction, which constitutes 10 to 20 percent of the tar (average, 17 percent), most of the naphthalene is removed. Phenols and cresols (0.3 to 1.1 percent of the tar) are then removed (77). A schematic flowchart for the production of cresols from coal-tar acid oil is presented in Figure 4 (77).

TABLE XII

PRODUCERS OF SYNTHESIZED p-CRESOL IN THE UNITED STATES IN 1972

COMPANY	DIVISION	LOCATION	CAPACITY (lbs)	PROCESS
Hercules	Organics	Gibbstown, N.J.	6,000,000	oxidation of p-cymene
Koppers	Organic Materials	Follansbee, W.Va.	10,000,000	methylation of phenol
Continental Oil	Conoco Chemicals, Pitt-Consol Chemicals	Newark, N.J.	80,000,000	methylation of phenol
Sherwin-Williams	Sherwin-Williams Chemicals	Chicago, Ill.	10,000,000	sulfonation of toluene
		Total:	106,000,000	

Source: Data from U.S. Environmental Protection Agency, "Development Document for Proposed Effluent Limitations Guidelines and New Source Performance Standards for the Major Organic Products Segment of the Organic Chemicals Manufacturing Point Source Category," EPA 440/1-73/009, Washington, D.C., p. 210, December, 1973.

FIGURE 3
LOCATION OF PLANTS THAT SYNTHESIZE p-CRESOL

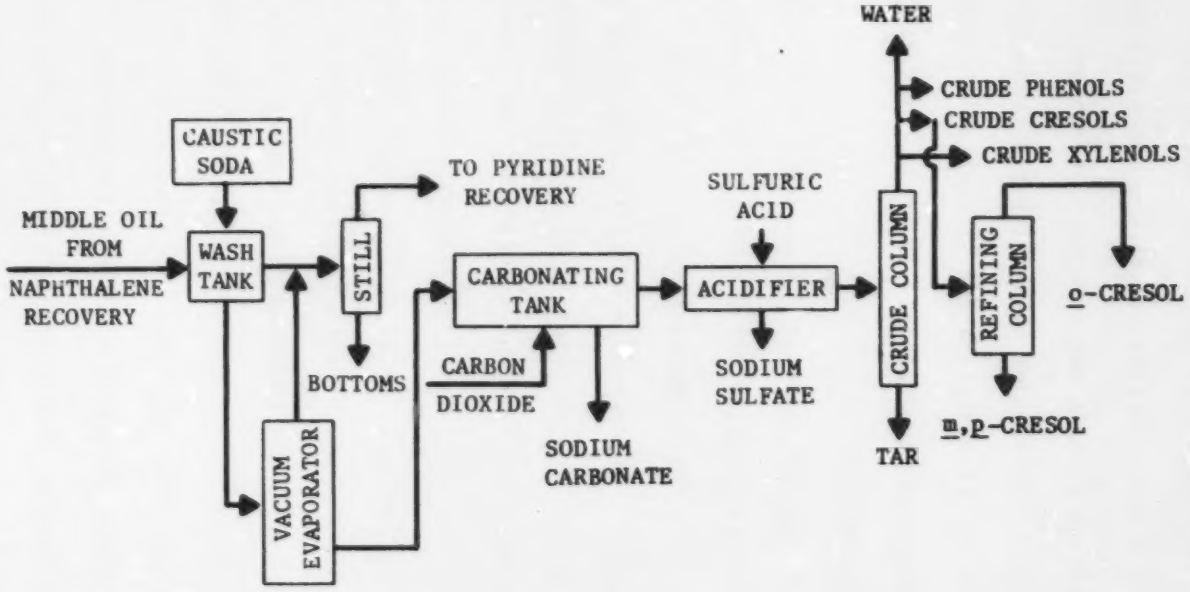
TABLE XIII

AVERAGE CONTENTS OF COAL TARS

TAR SOURCE	WEIGHT IN CRUDE TARS, PERCENT			
	TAR ACIDS	o-CRESOL	m-CRESOL	p-CRESOL
Coke oven	3	0.32	0.45	0.27
Continuous vertical retort	15	1.53	1.05	0.86
Low temperature	25	1.48	0.98	0.87

Source: From D. McNeil, "Cresols," in "Kirk-Othmer Encyclopedia of Chemical Technology," 2nd ed., Vol. 6, p. 437, Wiley, New York, 1965.

FIGURE 4
SCHEMATIC FLOWCHART FOR PRODUCTION OF CRESOLS FROM COAL TAR



Source: W. L. Faith, D. B. Keyes, and R. L. Clark, "Industrial Chemicals," 3rd Ed., John Wiley, New York, p. 292, 1965

In this process, the tar-acid oils are charged into a wash tank where the phenols are extracted with sodium hydroxide solution. Caustic concentration may vary from 8 to 50 percent, but higher concentrations are generally used. The tar acids are converted into their respective salts, sodium phenate and cresylate, which are soluble in the aqueous layer. This layer containing the tar-acid salts is allowed to settle in the wash tank and is then discharged from the bottom. The nonaqueous (neutral-oil) layer is distilled and sent to a pyridine-recovery plant.

Sodium cresylate from the wash tank is freed of entrained and dissolved oils and bases by evaporation under vacuum. The concentrated cresylate solution is run from the evaporator into a neutralizer or carbonating tank where most of the salts are converted into free tar acids by treatment with carbon dioxide. The acids are decanted from the resulting sodium carbonate solution and are then charged into an acidifier. Sulfuric acid, in varying concentrations, is added to complete the process of "springing" the tar acids. Some plants use sulfuric acid entirely, eliminating the carbon dioxide treatment, but this is not the usual practice. The released phenols and cresols (tar acids) are separated from the aqueous sodium sulfate solution in the acidifier by decantation. The top layer is distilled either intermittently or continuously through a fractionating column. The water is removed, and several fractions such as crude phenols, cresols, and xylenols are withdrawn. The crude cresol cut is further purified by fractional distillation to yield o-cresol and a mixture

consisting of m- and p-cresol. The latter cresols are generally marketed as a mixture since their separation is considerably more difficult than the isolation of o-cresol. The two isomers (meta-para) may be refined by the formation of the oxalic acid addition product or by alkylation with an olefin (isobutylene) to yield compounds that are more readily separated. Hydrolysis and dealkylation, respectively, yield refined m- and p-cresols (72).

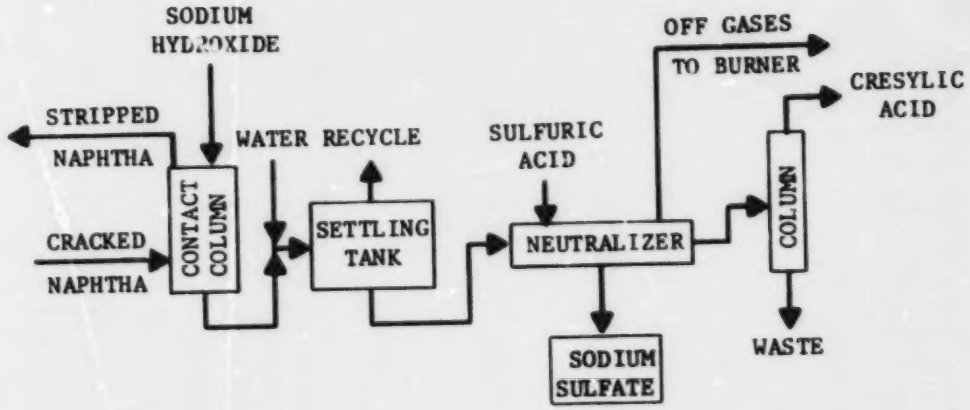
During the production of cresol from coal tar, cresols and cresylics might be emitted to the air as well as discharged with the waste water.

b. Production of Cresol from Petroleum

Petroleum-based cresol is derived from the petroleum acids produced during thermal or catalytic cracking of petroleum. A schematic flowchart of the production of cresylic acid from cracked naphtha is presented in Figure 5 (77).

In this process, cracked naphtha is passed through a vertical, packed contact column countercurrent to a solution of sodium hydroxide (caustic soda). A solution of sodium cresylate forms, and it is drawn off the bottom of the contactor. After being diluted with water, the solution is run to a setting tank, where it is allowed to stand until two layers separate. The hydrocarbon oil layer is drawn off the top and recycled. When a higher grade product is desired, the sodium cresylate solution may be steamed, filtered, and then solvent-extracted to remove additional hydrocarbon impurities. The dilute sodium cresylate solution is charged in batches into a gas-tight,

FIGURE 5
SCHEMATIC FLOWCHART FOR PRODUCTION OF CRESYLIC ACID FROM PETROLEUM



Source: W. L. Faith, D. B. Keyes, and R. L. Clark, "Industrial Chemicals," 3rd Ed., John Wiley, New York, p. 294, 1965

air-agitated reactor where it is neutralized by the addition of sulfuric acid. The gases formed are piped to a furnace, where they are burned before being discharged to the atmosphere. The sodium sulfate solution, which is obtained in the neutralization step, separates, and it is discharged from the bottom of the new reactor to waste. Crude cresylic acid is drawn off the top of the reactor and run to a batch distillation column. The column operates under increasing vacuum as the distillation progresses in order to maintain low distillation temperatures. Various cuts of cresylic acid are taken, ranging from 350 to 525°F. The ortho isomer and meta-para isomer mixture are obtained. The grades are adjusted by blending to meet customers' specifications (77).

Petroleum-based cresol is also derived from spent caustic liquor. This liquor was used in oil refineries to scrub catalytically mercaptans from cracked gasoline. These waste liquors contain 25 percent phenols, cresols, and xylenols (77). Tar acids are separated from the liquor by various oxidation, decomposition, and extraction procedures (4). The recovered tar acids contain 49 percent cresols (18 percent o-cresol, 22 percent m-cresol, and 9 percent p-cresol) as well as phenol, xylenols, and higher phenolics. The o-cresol and m-,p-cresol fractions are separated by distillation of the cresylic acid layer.

Cresols may be emitted to the atmosphere during the production of petroleum-derived cresol. Cresols may also be present in the waste water discharges from various extraction and distillation processes.

c. Synthesis of Cresol

Cresols can be synthesized by sulfonation and hydrolysis of toluene, by methylation of phenol, and by oxidation and cleavage of cymene.

p-Cresol was the first cresol isomer to be synthesized commercially (70). It is synthesized from toluene by sulfonation and subsequent hydrolysis (70, 78, 79). The overall reactions are:

$$C_6H_5CH_3 + H_2SO_4 \quad (SO_3H)C_6H_4CH_3$$

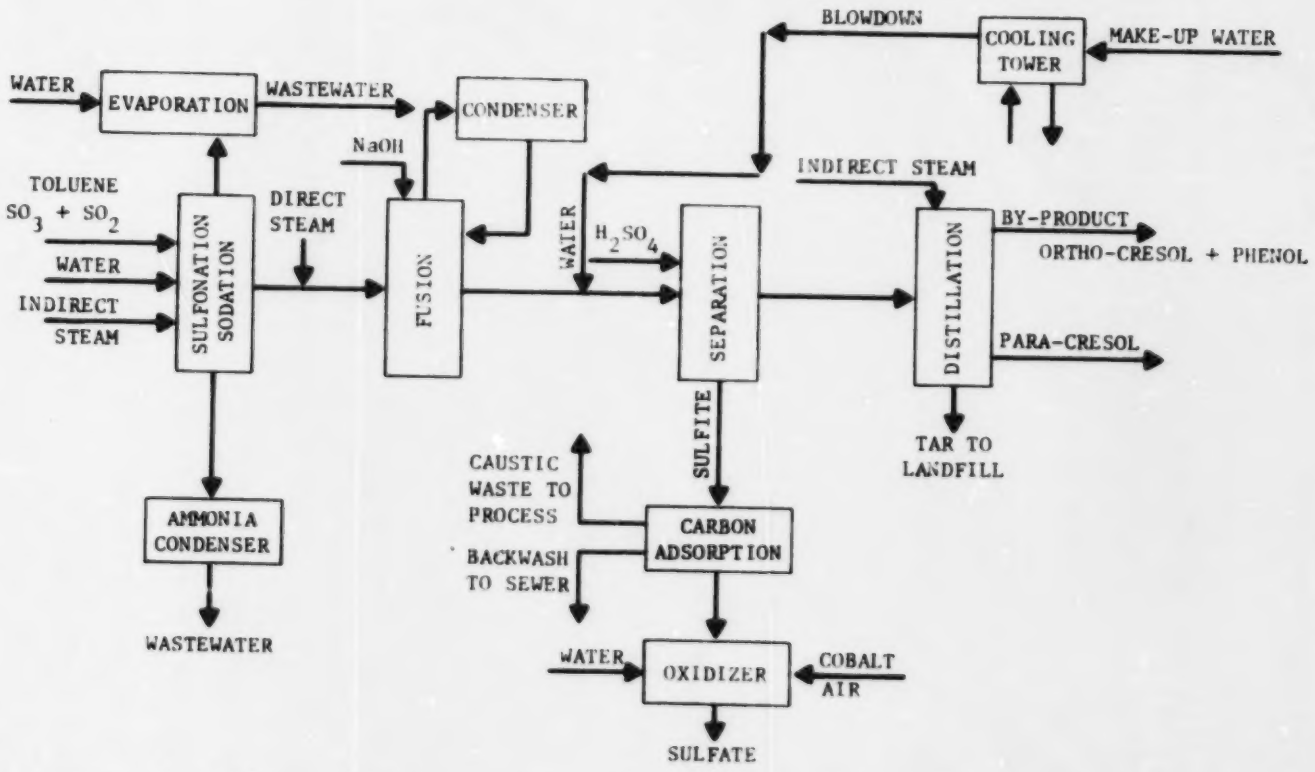
$$(SO_3H)C_6H_4CH_3 + NaOH \quad (OH)C_6H_4CH_3 + Na_2SO_3$$

A schematic flowchart for this process is presented in Figure 6 (70).

A study was made of the effluent discharged by a plant that synthesizes p-cresol by sulfonation of toluene (70). The major waste water stream was the aqueous phase discharged from the washing-separation column. This stream contained 2.1 percent organic compounds such as cresylics, cresols, and phenols at a concentration of about 10 mg/ml. This amounts to a loss to the water of about 1.1 pounds of cresylics, cresols, and phenols per 1000 pounds of p-cresol synthesized, or 0.11 percent.

If it is assumed that during synthesis there is a 10 percent loss in yield to the air and that pollution control devices are 98 percent effective, then losses would amount to 0.2 percent. Therefore, production of the 10 million pounds of p-cresol synthesized from toluene in the United States in 1972 would have been the source of

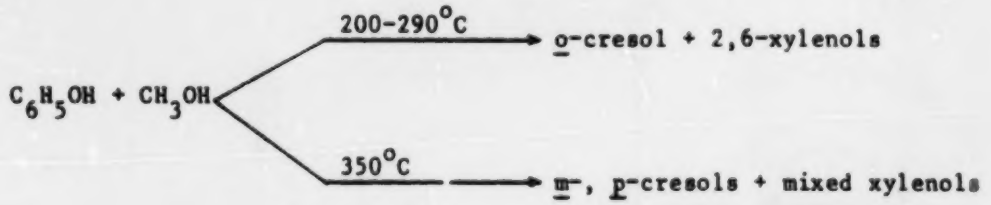
FIGURE 6
SCHEMATIC FLOWCHART FOR THE SYNTHESIS OF p-CRESOL FROM TOLUENE



Source: U. S. Environmental Protection Agency, "Development Document for Proposed Effluent Limitations Guidelines and New Source Performance Standards for the Major Organic Products Segment of the Organic Chemicals Manufacturing Point Source Category," EPA 440/1-73/009, Washington, D. C., p. 208, Dec. 1973

20,000 pounds of cresylics, cresols, and phenols emitted to the atmosphere.

The meta and para isomers of cresol are produced by vapor-phase methylation of phenol over porous alumina or silica-alumina catalyst (70, 78). o-Cresol is produced similarly at lower temperatures (78). The reactions are:



p-Cresol and m-cresol are also synthesized from p- and m-cymene, respectively (70, 78). Cymene is readily available as a by-product of paper pulp manufacture (78). The reactions, oxidation and then cleavage, are:

$$(CH_3)_2CHC_6H_4CH_3 + O_2 \xrightarrow{H+} (CH_3)_2CHC_6H_4OOH$$

$$(CH_3)_2CHC_6H_4OOH \longrightarrow CH_3C_6H_4OH + (CH_3)_2CO$$

In 1972, total United States capacity for synthesis of p-cresol was 106 million pounds (see Table XII). If, as discussed above, losses to the air are assumed to be 0.2 percent, then atmospheric emissions of cresol that result from p-cresol synthesis would have amounted to 212,000 pounds during 1972. If it is assumed that the 0.11 percent loss to water during p-cresol synthesis from toluene is

characteristic of all the synthetic processes, then cresol emissions in the waste water would have totaled 116,600 pounds in 1972.

2. Production of Coke

Coke production, from which cresol is a by-product, is a possible source of cresol emissions to the atmosphere. In 1974, coke production in the United States amounted to 62,256,066 tons (72). The 14.5 gallons of tar produced with each ton of coke would contain about 0.15 gallons of cresols (4, 77). From the hydrocarbon emissions that have been estimated for coke ovens with no emission controls (73), it is estimated that under these conditions 0.047 pounds of cresols are emitted to the atmosphere for each ton of coke produced. Therefore, if it is assumed that no significant emission controls were applied to coke ovens, approximately 2.95 million pounds of cresols were released to the atmosphere during coke production in the United States in 1974.

3. Use of Cresol

Table XIV lists the quantities of cresol that have been produced from natural sources, synthesized, and imported into the United States in recent years. These figures reflect increasing demand for cresols. The specifications for commercial cresols are listed in Table IV (3). The cresol isomers are used in many products; see Table XV (3). Table XVI (74, 80, 81) provides an indication of the apportionment of cresol consumption.

In 1973, the United States demand for p-cresol was 17,000 tons (82). Its main use was in the manufacture of 2,6-di-tert-butyl-p-cresol

TABLE XIV

CRESOL MARKET IN THE UNITED STATES (MILLIONS OF POUNDS)

YEAR	PRODUCTION		IMPORTS*	REF.
	REFINED	SYNTHESIZED		
1968	152	3	64.5	74
1969	171.2		14.4	75
1970	189.7	98**	7.9	75
1971	151.1***		5.3	
1972		106		70
1973	115.4			71

*Import data are for crude cresylic acid (75).
**Production of cresylic acid (78).
***Estimated (75).

TABLE XV
USES OF CRESOLS

Commercial Cresol	o-Cresol	m-Cresol	p-Cresol	Cresylic Acid
phenolic resins tricresyl phosphate disinfectants surfactants flotation agents scouring compounds chemical intermediates lube oil additives enamel solvent	disinfectant coumarin organic intermediate	disinfectant fumigating compositions production of synthetic resins photographic developer nitrocresol explosives ore flotation intermediate ink, paint, and varnish remover reclaiming rubber	disinfectant fumigating compositions cresotinic acid dyestuffs organic intermediate for: antioxidants resins pharmaceuticals dyes pigments	petroleum derived: tricresyl phosphate disinfectants metal cleaning compounds phenolic resins flotation agents surfactants chemical intermediates oil additives coal tar derived: phenolic resins wire enamel solvent tricresyl phosphate solvent refining of lubricating oils disinfectants scouring compounds detergents pesticides

*In order of volume.

Source: Data from A. Rose and E. Rose, "The Condensed Chemical Dictionary," 7th ed., Van Nostrand Reinhold, New York, pp. 257-259, 1966.

TABLE XVI

USES OF CRESOLS AND CRESYLIC ACIDS IN THE UNITED STATES
(Millions of Pounds)

	1957	1968	1973
Phosphate esters	30.0	63	39.6
Magnet wire			27.0
Antioxidants			27.0
Resins	33.6	50	27.0
Exports	7.3		18.0
Cleaning and disinfectant compounds			10.8
Engine, metal cleaners	9.8		
Disinfectants	12.5		
Textiles, medicinals, dyes, inks	10.4		
Ore flotation reagents	8.3		10.8
Wire enamel solvent	7.2	17	
Miscellaneous	4.7	25*	19.8
Total	123.8	155	180

*Includes disinfectants, exports, salicylaldehydes, herbicides, ore flotation reagents, and metal cleaners.

Sources: Data for 1957: Oil Drug Paint Reporter, Sept. 29, 1958;
data for 1968: Chem. Eng. News, May 20, 1968, p. 24;
estimate for 1973: Chem. Mark. Rep., July 23, 1973.

(BHT), a general purpose antioxidant (80) that is used as a food additive, in polymers, and in elastomers (83). The use of p-cresol in fragrances is less than 2000 pounds annually in the United States (55). p-Cresol is also used to produce oil-soluble resins (4).

o-Cresol is used mostly in the manufacture of the herbicides 2,4-dinitro-o-cresol (DNOC) and 2-methyl-4-chlorophenoxyacetic acid (MCPA) (4).

The m-cresol isomer is used in the production of plastic molding compositions and adhesives (4).

Cresylic acid and the m-,p-cresol mixtures are used in the manufacture of resins and of tricresyl phosphates (TCP). TCP is used as a plasticizer for poly(vinyl chloride) and also as an additive to gasoline and hydraulic fluids (4, 84).

Cresols and cresylic acids are used as disinfectants. They are usually marketed as stabilized saponated solutions (2, 4). These products are widely used in households (6).

4. Disposal

Disposal of cresols and cresol-containing products as well as their containers is a potential source of environmental loading. About 90 percent of the solid wastes in the United States are deposited in landfills and 10 percent are disposed of by incineration (85).

Unless the containers are, and remain, sealed, cresols can leach from the landfills. The hydrosphere (the ground water and/or the surface water) will then be polluted with cresol.

Cresols would be vaporized in most incinerators because the boiling point of the cresol isomers ranges from 191 to 203°C, temperatures which are below the operating temperatures of most municipal incinerators. Cresols are slightly flammable, and cresol vapor that is exposed to heat or flame is explosive; furthermore, cresols are decomposed by heat. It is therefore unlikely that incineration is a source of cresol emissions to the atmosphere.

E. CONTROL

Adsorption onto activated carbon is the best method for controlling emissions of cresols. Activated carbon has a high capacity for cresol; one pound adsorbs about 0.2 to 0.5 pounds of cresol, the average being about 0.33 pounds (86). Another advantage of this method is that the compounds can be recovered when the spent carbon is regenerated.

In a demonstration plant, activated carbon was used to remove organic compounds from the waste water of a plant that synthesized p-cresol. It was reported that the influent contained 3500 to 6500 milligrams of cresylic compounds per liter, whereas the concentration in the effluent ranged from 0 to 700 mg/l. The spent carbon was regenerated with 10 percent sodium hydroxide solution, and the recovered cresylic compounds were recycled back into the process (70).

Adsorption onto activated carbon can also be used to control cresol emissions to the atmosphere. This method is feasible where the air streams from ventilating systems can be directed through activated carbon in a vapor-phase adsorbent system. Activated carbon adsorption is applicable to any type of facility, and it is irrelevant whether the cresol emissions derive from production, purification, or processing of cresols.

Another method of controlling atmospheric emissions of cresol is incineration of the vapors. Off-gases are diverted to a furnace where they are incinerated before being discharged to the atmosphere. This control technique is used with the production of petroleum-derived cresol (77).

Cresol usually does not present an acute inhalation hazard because of its low vapor pressure and its odor detectability at concentrations well below those considered dangerous. The United States Occupational Standard, a maximum time-weighted average concentration that is based on a 40-hour work week, is 5 ppm for total cresol (19, 52, 53, 87) or 22 milligrams of cresol per cubic meter of air (87). It should be noted, however, that the ambient air quality standards for cresol in the Federal Republic of Germany are a basic standard (i.e., for long-term exposure) of only 0.05 ppm or 0.2 mg/m^3 (STP) for a 30-minute averaging time and a permissible standard (i.e., not to be exceeded more than once in any four hours) of 0.15 ppm or 0.6 mg/m^3 (STP) (88). Thus the United States TWA for cresol is ten-fold the concentration permitted in Germany.

According to the Chemical Safety Data Sheet published by the Manufacturing Chemists Association (62), special ventilation is usually not required in industry. However, it may be necessary where hot cresol vapor is emitted in an operation, and such ventilation systems should be designed to provide downdraft mechanical exhaust because cresol vapor is 3.72 times heavier than air and consequently settles. Ventilation systems that are designed for cresol vapors should certainly be vented through a pollution control device (e.g., an activated carbon adsorption system) before the air is released to the atmosphere.

Procedures which might be classified as general good housekeeping techniques will also serve to reduce cresol exposure to air. These include the use of closed systems whenever and wherever possible, frequent equipment inspections to detect and to prevent leaks, and immediate cleanup of leaks and spills (e.g., by flushing with water, by absorbing with sawdust or commercial oil absorbents). By limiting the exposure of cresol to air and, consequently, its vaporization, these practices will help to curtail the emission of cresol to the atmosphere.

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